Determination of Heavy Element Nanophases Embedded in Vitreous Silica via Complementary X-ray and Neutron Scattering Techniques

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Using the BESSRC 12-ID-C beam line at APS, small angle X-ray scattering (SAXS) studies were carried out as a function of X-ray energy near the L_3 absorption edge of heavy element ions in phosphate phases that were embedded in vitreous silica. The resulting anomalous small angle X-ray scattering (ASAXS) measurements conclusively showed that the heavy element ions in these materials are present in nanophases. Analysis of the data for X-ray energies far below the absorption edge determined the size of the nanophases. Using ASAXS on a series of samples each of which was doped with a single heavy element, nanophases were identified that contained ions of the heavy elements europium, gadolinium, terbium, and lutetium. Our previous small angle neutron scattering (SANS) work at IPNS on the terbium-containing material had identified the presence of nanophases in it and determined their size. To within experimental error, the nanophase size determined by SAXS and SANS were identical. Our studies have shown that ASAXS and SANS are complementary techniques. ASAXS quickly and directly identifies nanophases that contain a particular heavy element in a sample volume of 5 millionths of a cubic centimeter whereas SANS provides quantitative measurements over macroscopic sample volumes for contained nanophases whose density differs, in our case, from that of silica. The materials that we are investigating are part of our development of a single approach to reducing nuclear waste volume in a Nuclear Energy Research Initiative (NERI) project. The ability of ASAXS to investigate gadolinium phases is of particular importance because gadolinium is the element of choice for criticality control in nuclear waste that contains fissile isotopes. Due to the large neutron absorption cross-section of gadolinium, however, SANS studies on gadolinium phases are impractical. We also are investigating the unusual photophysical properties of these heavy element materials in work funded by the Division of Chemical Sciences of the Office of Basic Energy Sciences. Our research team for these ASAXS and SANS studies included S. Skanthakumar and Soenke Seifert from the Chemistry Division and Lixin Fan and P. Thiyagarajan from the Intense Pulsed Neutron Source Division.